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Gas hydrate of Lake Baikal: Discovery and varieties

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1. Introduction

Lake Baikal is the largest and the oldest fresh-water reservoir on our planet. The lake basin is located in Eastern Siberia on the border between the Siberian Craton and the Central Asian Fold Belt, and it forms the central part of the large, intracontinental Baikal Rift Zone. The lake is more than 600 km long, 80 km wide (in the widest place) and its water depth amounts up to 1637 m at deepest point. With a total basement subsidence of ~ 10 km, more than $\frac{3}{4}$ of this huge depression are filled up with several kilometers of sediments. Their total volume is estimated to be in the order of 75,000 km³ (Logachev, 2003). The age of the oldest sedimentary deposits in the Central and the Southern Basins of the lake surpasses 25 Ma (Oligocene), and there are some data testifying that the sediments in the Central Basin, near the delta of Selenga River, started to accumulate as far back as in Palaeogene (ca. 60 Ma) (Nikolayev, 1998). Geophysical data indeed reveal a maximum thickness of the sedimentary infill (about 9 km) near the Selenga delta (Scholz and Hutchinson, 2000). Elsewhere in the Southern and the Central Basin, the sedimentary thickness does not exceed 7.5 km, while in the north-east of the Northern Basin it amounts to 4.4 km (Hutchinson et al., 1991). Such a long sedimentation history and such large amounts of sediments containing a considerable amount

ABSTRACT

This paper summarizes the results of recent gas-hydrate studies in Lake Baikal, the only fresh-water lake in the world containing gas hydrates in its sedimentary infill. We provide a historical overview of the different investigations and discoveries and highlight some recent breakthroughs in our understanding of the Baikal hydrate system. So far, 21 sites of gas hydrate occurrence have been discovered. Gas hydrates are of structures I and II, which are of thermogenic, microbial, and mixed origin. At the 15 sites, gas hydrates were found in mud volcanoes, and the rest six - near gas discharges. Additionally, depending on type of discharge and gas hydrate structure, they were visually different. Investigations using MIR submersibles allowed finding of gas hydrates at the bottom surface of Lake Baikal at the three sites.

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of organic matter favoured the formation and accumulation of different types of hydrocarbons. These oils and natural gases migrate in free or dissolved state towards the lake floor where high pressures (corresponding with the large water depth) and low temperatures of the near-bottom waters (3.5 °C approximately) are propitious for the formation of gas hydrates.

2. History and results of study on Baikal gas hydrates

The history of gas hydrate investigations in Baikal can be subdivided in four stages with key moments in 1997, 2002, and 2008.

Before 1997, the possible presence of gas hydrates was evoked in a few reports. The very first reference to hydrates in Baikal was made in a VNIIGAZ study, indicating a "site with possible gas hydrate accumulation" in sediments of Lake Baikal (Yefremova et al., 1980). The discovery of the first Bottom-Simulating Reflector (BSR) during a multi-channel seismic survey in 1989 (Hutchinson et al., 1991) was the first real indirect geophysical indication for the presence of hydrates. Later, a predictive map depicting the base of the hydrate stability zone in Lake Baikal was published, which was based on geothermal modeling and heat-flow measurements (Golubev, 1997). A more precise map of the base of gas-hydrate layer corresponding to the mapped BSR recognized on multi-channel seismic data from 1992 was published by Golmshtok et al. (1997). Interestingly, the BSR was only observed in the Southern and Central Basin and was not always parallel to the lake floor. These first estimates of the depth and extent of the gas hydrate



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Fig. 1. Geographical distribution of the sites where gas hydrates have been studied in Lake Baikal. The square indicates the location of the BDP-97 borehole; circles show mud volcanoes (mv) or the sites of underwater discharge of gas or gas-saturated fluids (seep).

stability zone were used to make the first assessments of the resource potential of the Baikal hydrate reservoir yielding estimates varying from 8.8×10^{11} to 9×10^{12} m³ (of gas at STP conditions) (Golubev, 2000; Vanneste et al., 2001). A key discovery was made in 1997 when the first and only samples of deep gas hydrates were obtained during the International Baikal Drilling Project (BDP). While drilling the borehole BDP-97 from the ice, core samples taken from sub-bottom depths of 121 and 161 m contained frozen sand-silt material that released abundant amounts of gas while heated. Laboratory analyses of the sediment samples showed that the cement was, in fact, gas hydrate of cubic structure I (CS-I), composed mainly of methane of biogenic origin (CH₄ × 6H₂O; δ^{13} C between -58 and -68%) (Kuz'min et al., 1998).

From 1997 onwards, gas-hydrate research on Lake Baikal began to focus on hydrate occurrences at or near the lake bottom. Between 1997 and 2002, different geophysical studies were carried out in various regions of the Southern and the Central Basins (De Batist et al., 2002), aimed at mapping and characterizing a series of lake-floor structures related to the discharge of gas and/or gas-saturated water. Earlier, Ginsburg and Solovyev (1994) had established that the main deposits of near-bottom submarine gas hydrates in oceans and seas occur in association with such seep structures. The new seismic, side-scan sonar and bathymetric data revealed four such seep structures along one of the main faults within the Southern Basin. On the side-scan sonar mosaics, these structures ranged from 200 to 2000 m in diameter and up to 60 m in height, had practically isometric contours in the center of which one or more culminations could be seen; the echograms outlined positive topographic structures. The seismic data showed below these structures "muted" (transparent) acoustic signals and an irregular BSR behavior. Owing to their size and morphological characteristics they were named "Bol'shoy" (Big), "Malen'kiy" (Small), "Malyutka" (Baby) and "Stariy" (Old) (Van Rensbergen et al., 2002; De Batist et al., 2002). One year later, in 2000, coring of "Malen'kiy" structure allowed for the first time the recovery of surface (near-bottom) gas hydrates in the form of lenses, strata, and massive formations within clayey sediments (Klerkx et al., 2003; Matveeva et al., 2003). Hydrate and gas analyses demonstrated that these were CS-I methane hydrates of biogenic origin.

From 2002 to 2008, geological and geophysical studies focused further on the search for new sites of hydrocarbon discharge with gas hydrate occurrences. Geophysical data, acquired in 2002 in the Central Basin, revealed four new structures in Kukuy Canyon, which were named "K-1","K- 2", "K-3" and "K-4", and two – in the vicinity of Olkhon Island, which were named "Saint-Petersburg" and "Novosibirsk" (Klerkx et al., 2006) (Fig. 1). In 2003 and 2004, hydrates were taken from the "Bol'shoy" and "K-2" structures. Similar to the observations at the "Malen'kiy" structure, the hydrate-bearing sediment in the "Bol'shoy" and "K-2" structures consisted of a mud breccia, which was clearly different from the typical alternating diatomaceous and clayey layers normally found in the surface sediments of Lake Baikal. Such type of breccia



Fig. 2. Photos of different types and textures of gas hydrates in Lake Baikal. (A) pseudoporphyritic; (B and C) massive; (D) granules; (E) granules and vertical layers/veins; (F) layers and cement-like hydrate between them (a gray sludge between white layers); (G) layers; (H–J) veins and streaks of different occurrence.

has become a geological search indicator of hydrates in sediments of the lake (Khlystov, 2006). The occurrence of hydrates in these seep structures was also indicated by the presence of strong reflectors on very-high-resolution sub-bottom profiler data (5 kHz). These strong reflectors correlate exactly with the locations of gas hydrate sampling, thus, making them a geophysical search indicator (Khlystov et al., 2007). More gas-hydrate-bearing structures were discovered in 2005, 2007 and 2008 during several side-scan sonar and acoustic profiling surveys: the "P-1" and "P-2" structures opposite Peschanaya Bay (Southern Basin), the "K-0", "K-5", and "K-6" structures in the vicinity of Kukuy Canyon and new gas seeps "G-1", "G-2", and "G-3" in the area of Bolshoye Goloustnoye and oil-gas seep near cap "Gorevoy Utes" (see Fig. 1). The geological and geophysical data showed that most of these hydrate-bearing structures, like the ones discovered in earlier years, share many characteristics with mud volcanoes (Khlystov et al., 2007; Hachikubo et al., 2010).

After 2008, the survey using multibeam echosounder and deepsea manned submersibles MIR-1 and MIR-2 played a major role in search for new sites of gas hydrate accumulations.

In 2008–2010, several dives of MIR submersibles in Baikal allowed detailed visual exploration of the identified seep and mud volcano structures. During these dives, small bottom structures consisting of hydrate and bitumen were found at the bottom surface of the oil seep "Gorevoy Utes" (at 900 m water depth) (Khlystov et al., 2009) (cf. video surveillance in Supplementary material); gas hydrate layers of several meters in thickness were found on the flanks of the seep "Saint-Petersburg", at (1400 m water depth) (Egorov et al., 2009); and two gas hydrate layers of less than one meter in thickness were found in the wall of the canyon in the seep area "Goloustnoe", (at 400 m water depth) (cf. video surveillance in Supplementary material).

Data obtained in 2008–2010 using multibeam echosounder allowed discoveries of 11 new sites with gas hydrate accumulations near bottom surface of the lake. As before, most of these accumulations are associated with mud volcanoes, which are well defined in the bottom topography (mud volcanoes "K-1", "K-4", "K-8", "K-9", "K-10", "K-11", "Krest", "Novosibirsk") and some – with gas discharges (seep "13", "Saint-Petersburg", "K-12"); however, there were also some accumulations at the sites neither with defined bottom topography, nor with evidence of gas discharges. For instance, during dives of MIR submersibles at a depth of 500 m, vast fields of bottom surface sediments of different color with gas hydrate accumulations under them were found on the slope of underwater high Posolskaya Bank (seep "PosolBank", see Fig. 1). In any case, all these sites are located above discontinuous faults cleaving bottom sediments to the depths of BSR occurrence (350–400 m).

3. Gas hydrate varieties

In most cases, dissolved gas and hydrate gas contained 98% methane (C₁) of bacterial origin. Hydrates containing thermogenic ethane (C₂) were first found at the "K-2" structure, and thermogenic methane was later found at Gorevoy Utes (Kalmychkov et al., 2006). Moreover, it was found that hydrates of cubic structure II (CS-II) existed in the "K-2" mud volcano (Kida et al., 2006; Hachikubo et al., 2010). In 2010-2011, we have discovered such hydrates at two more mud volcanoes of the Kukuy region, mud volcanoes "K-10" and "K-4" with high thermogenic methane content relative to methane. In 2010, similar hydrates were found in the Southern Basin of the lake on the slope of underwater high Posolskaya Bank, seep "PosolBank". X-ray diffraction study of this sample was performed using synchrotron radiation at the 4th beamline of the VEK-PP-3 storage ring (Siberian synchrotron and terahertz radiation center, Budker Institute of Nuclear Physics SB RAS) (Ancharov et al., 2001). Analysis of the diffraction patterns has shown that the sample contain hydrate of structure II. Table 1 shows the data on all gas hydrates in Lake Baikal discovered since 2000, including their geometries, content, genesis, and structure types.

The near-surface hydrates in Lake Baikal occur over a waterdepth range of more 1 km (from \sim 1430 m in the "Novosibirsk" mud vulcano to \sim 390 m in the seep "G-2") and the hydrates show wide variety of textures, including pseudoporphyritic, massive, layered and vein structures (Fig. 2). In a single instance, hydrate in the form of cement was obtained from the "Malen'kiy" mud

Table 1

Characteristics of gas hydrates in Lake Baikal.

No.	Name	Location, depth	Discovery year	Hydrate form (texture)	Average relationship C_1/C_2	Type of gas	Type of structure
1	mv Malen'kiy	51.9274°N 105.6486°E	2000	Pseudo-porphyritic, layers, cement-like	5272	Microbial	CS-1
		1280 m 51 8771°N		hydrate			
2	mv Bol'shoy	105.5490°E	2003	Massive, pseudoporphyritic, layers, veins and streaks	4352	Microbial	CS-1
3	mv K-2	1380 m 52.5895°N 106 7692°E	2005	Granules	6	Mixed	(5-2
		900 m 52.5026°N	2000	Vertical layers/veins	29		
4	mv K-0	106.6111°E 415 m 51.9076°N	2006	Veins and streaks	1002	Microbial	CS-1
5	mv Maluytka	105.6032°E 1307 m	2006	Layers, veins and streaks	4840	Microbial	CS-1
6	mv K-6	52.6001°N 106.8149°E	2007	Massive, pseudoporphyritic, layers, veins and streaks	2849	Microbial	CS-1
7	my Deschanka (D-2)	1005 m 52.1737°N 105 8097°F	2007	Massive layers yeins and streaks	7200	Microbial	CS-1
,		825 m	2007		7200	WICIODIAI	CJ-1
8	Seep Goloustnoe (G 1-3)	51.9762°N	2008	Layers, veins and streaks	297	Mixed	CS-1
		105.3493°E 410 m	2010	Massive layers on bottom			
9	Oil seep Gorevoy Utes	53.3013°N 108.3890°E 875 m	2006 2008	Massive, layers, veins and streaks Massive with bitumen on bottom	584	Thermogenic	CS-1
10	Seep Saint- Petersburg	52.8824°N 107.1675°E 1402 m 52.0360°N	2009	Massive hall, layers on bottom	300	Mixed	CS-1
11	Seep PosolBank	105.8433°E 500 m	2010	Granules Layers/veins	6 32	Mixed	CS-2
12	mv K-1	52.5667°N 106.7130°E 675 m	2010	Layers, veins and streaks	217	Mixed	CS-1
13	mv K-9	52.5879°N 106.7251°E 741 m	2010	Massive, layers, veins and streaks	7572	Microbial	CS-1
14	mv K-10	52.5325°N 106.6550E 531 m	2010	Small granules Layers, veins and streaks	6 31	Mixed	CS-2 CS-1
15	mv Novosibirsk	52.9348°N 107.2548°E 1396 m	2010	Massive, layers, veins and streaks	290	Mixed	CS-1
16	mv K-8	52.5195°N 106.6630°E 425 m	2010	Layers, veins and streaks	78	Mixed	CS-1
17	mv K-11	52.5171°N 106.6771°E 419 m	2011	Layers, veins and streaks	37	Mixed	CS-1
18	Seep K-12	52.5047°N 106.5882°E 489 m	2011	Layers and streaks	1848	Microbial	CS-1
19	mv K-4	52.5883°N 106.7498°E 835 m	2011	Granules Layers, veins and streaks	6 35	Thermogenic	CS-2 CS-1
20	Seep 13	52.9338°N 106.8691°E 1134 m	2011	Layers and streaks	75	Mixed	CS-1
21	mv Krest	52.9601°N 106.9438°E 1280 m	2011	Massive, layers, veins and streaks,	49	Mixed	CS-1

volcano. In the mud volcanoes "K-2" and "K-4" hydrates occurred both as vertical veins and in the form of granules (Fig. 2). Both types were found in the same cores, either directly in contact with each other or separated by an erosive surface. Granular gas hydrates were also found in bottom sediments of the seep "PosolBank".

4. Discussion and conclusions

The structural types of the most common gas hydrates (structures I and II. structure H) were considered by Sloan and Koh (2008). Hydrates of structure I formed by methane with a small admixture of heavier hydrocarbons are the most widespread. Methane mixed either with propane (more than 0.2 mol.%) or ethane (2–22 mol.%) forms hydrates of structure II. For example, this type of hydrates is often formed by associated oil gas. In addition, gas mixtures containing methane and higher hydrocarbons (methyl cyclohexane as an example) can form hydrates of structure H that are rare in nature. Thus, hydrates of the three structures were found in the same accumulation GC 234 (Northern Gulf of Mexico) (Yousuf et al., 2004). Although search of such structures was also carried out in Baikal, including oil seep with gas containing heavy hydrocarbons, we succeeded in finding and identification by laboratory methods of only two types of structures (i.e. CS-I and CS-II). Samples of coexisting hydrates of different cubic structures (CS-I and CS-II) composed of mixed microbial methane and deep thermogenic methane and ethane were obtained from the mud volcano "K-2", "K-4", "K-10", and the seep "PosolsBank".

Therefore, using advanced geophysical equipment with high resolution, deep-sea manned submersibles MIR and developed geological and geophysical search indicators of gas hydrates in Lake Baikal, four mud volcano sites with 18 mud volcanoes have been discovered so far on the lake floor. Subsurface gas hydrates of different structures were taken from the sediments of 15 mud volcanoes and six seep fields. Three of them were found to have either gas hydrate outcrops or gas hydrate and bitumen mounts at the bottom surface of Lake Baikal.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.jseaes. 2012.03.009.

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